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Abstract

A dynamical theory that connects electronic motions and the nonlinear optical response of conjugated polyenes is developed by introducing the concept of electronic normal modes. A novel picture for the mechanism of optical nonlinearities is obtained by identifying the few dominant modes. This quasiparticle electron-hole representation establishes a close analogy with small semiconductor particles (quantum dots), and is very different from the traditional approach based on the electronic eigenstates. The effective conjugation length (coherence size), which controls the scaling and saturation of the static third order susceptibility $\chi^{(3)}$ with the number of double bonds, is related to the coherence of the relative motion of electron-hole pairs created upon optical excitation.

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Publications in Reviewed Journals

1. "Simulation of the Ultrafast Optical Response of Water", L. E. Fried and S. Mukamel, in Time-Resolved Vibrational Spectroscopy V, H. Takahashi Editor, Springer-Verlag, Berlin, (1992) pp. 295- 298.

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15. "Femtosecond Four Wave Mixing Spectroscopy of Conjugated Polymers", M. Hartmann and S. Mukamel, J. Chem. Phys., **99**, 1597-1606 (1993)

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University of Indiana, Department of Chemistry, "Femtosecond Optical Spectroscopy of Molecular Systems in Condensed Phases" Bloomington, Indiana, November 1992.

University of Pittsburgh, Department of Chemistry, "Nonlinear Femtosecond Spectroscopy of Molecular Systems in Condensed Phases", Pittsburgh, Pennsylvania, November 1992.

Joint Harvard - MIT Seminar, "Cooperative and Ultrafast Nonlinear Optical Response of Confined Excitons and Molecular Nanostructures", Cambridge, Massachusetts, December 1992.

SPIE International Symposium on Advanced Electronic and Optoelectronic Materials, "Nonlinear Optical Response of Conjugated Systems : Electron-Hole Anharmonic-Oscillator Picture," Los Angeles, California, January 1993.

ACS National Meeting, "Nonlinear Optical Response of Conjugated Polyenes," Denver, Colorado, March 1993.

CLEO/QELS Quantum Electronics and Laser Science Conference, "Third Order Nonlinear Spectra of a Polydiacetylene: The Quasi-Particle Exciton Representation", Baltimore, Maryland, May 1993.

French-German-Israeli Symposium on *Dynamical Processes in Condensed Molecular Systems*, "Cooperative Effects in the Nonlinear Optical Response of Confined Excitons," Garchy, France, May 1993.

Royal Netherlands Academy Colloquium on *Femtosecond Reaction Dynamics*, "Multimode Brownian Oscillator Analysis of Ultrafast Nonlinear Optical Response," Amsterdam, Netherlands, May 1993.

Sixth International Conference on *Time-Resolved Vibrational Spectroscopy*, "Two-Dimensional Off-Resonant Femtosecond Spectroscopy of Liquids", Berlin, Germany, May 1993.

Max-Planck-Gesellschaft zur Förderung der Wissenschaften, Arbeitsgruppe *Halbleitertheorie*, "Nonlinear Optical Response of Molecular and Semiconductor Nanostructures", Berlin, Germany, May 1993.

ETH, Laboratorium für Physikalische Chemie, "Optical Nonlinearities and Femtosecond Spectroscopy of Confined Excitons", Zürich, Switzerland, May 1993.

Canadian Society for Chemistry Conference, "Cooperative and Ultrafast Nonlinear Optical Response of Confined Excitons", Montreal, Canada, June 1993.

DPC '93, 9th International Conference on the Dynamical Processes in Excited States of Solids, "Retardation Effects in the Nonlinear Optical Response; Pump Probe Spectroscopy of Nanostructures, Cambridge, Massachusetts, August 1993.